

MESOSCOPIC PHASE SEPARATION IN THE MODEL WITH COMPETING JAHN-TELLER AND COULOMB INTERACTION.

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Abstract

We have derived the interaction between Jahn-Teller centers due to optical and acoustic phonons. Without Coulomb repulsion the model leads to a global phase separation at low temperature. At long distances Coulomb repulsion always dominates the short range attraction leading to phase separation on a short scale. On the basis of Monte-Carlo simulation of microscopic lattice-gas model we have formulated an effective phenomenological model. In the limit of low density of polarons, phase separation takes place in the form of charged bubbles. In the limit of high density, charge segregation occurs on domain walls.

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There is substantial evidence that the ground state in many of oxides is inhomogeneous[1]. In cuprates, for example neutron-scattering experiments suggest that phase segregation takes place in the form of stripes or short segments of stripes[2, 3]. There is some controversy whether this phase segregation is associated with magnetic interactions. On the other hand it is also generally accepted that the charge density in cuprates is not homogeneous.

The idea of charge segregation in cuprates appeared just after discovery of High-Tc materials [4, 5, 6]. In most cases long-range Coulomb repulsion was not considered. Recently it was suggested that interplay of the short range lattice attraction and the long-range Coulomb repulsion between charge carriers could lead to the formation of short metallic [7, 8] or insulating[8, 9] stripes of polarons. If the attractive potential is isotropic, charged bubbles have a spherical shape. Kugel and Khomskii [10] suggested recently that the anisotropic attraction forces caused by Jahn-Teller centers could lead to the phase segregation in the form of stripes. The long-range anisotropic attraction forces appear as the solution of the full set of elasticity equations (see ref.[11]). Alternative approach to take into account elasticity potentials was proposed in ref.[12] and is based on the proper consideration of compatibility constraint caused by absence of a dislocation in the solid.

Recently we formulated the model[13] where we suggested that interaction of a two-fold degenerate electronic state with τ_1 phonon modes at a finite wave-vector can lead to a local nonsymmetric deformation and short-length scale charge segregation. It was suggested [14] that this phase separation may lead to the insulator to superconductor phase transition governed by percolation. In this paper we reduce the proposed model to the lattice gas model and show that the model indeed displays phase separation, which may occur in the form of stripes or clusters depending on anisotropy of the short range attraction between localized carriers. We also generalize the model taking into account interaction of the Jahn-Teller centers via elasticity induced field.

We first derive a simplified lattice gas model. We show that the model without Coulomb repulsion displays a first order phase transition at a constant chemical potential. When the number of particles is fixed, the system is unstable with respect to the global phase separation below a certain critical temperature. In the presence of the Coulomb repulsion, global phase separation becomes unfavorable due to a large contribution to the energy from long range Coulomb interaction. The system shows mesoscopic phase separation where the size of charged regions is determined by the competition between the energy gain due to

ordering and energy cost due to breaking of the local charge neutrality. Since the short range attraction is anisotropic the phase separation may be in the form of short segments or/and stripes.

Let us begin by considering a simplified version of the JT model Hamiltonian [13], taking only the deformation of the B_{1g} symmetry:

$$H_{JT} = g \sum_{\mathbf{n}, \mathbf{l}} \sigma_{3, \mathbf{l}} f(\mathbf{n}) (b_{\mathbf{l}+\mathbf{n}}^\dagger + b_{\mathbf{l}+\mathbf{n}}), \quad (1)$$

here the Pauli matrix $\sigma_{3, \mathbf{l}}$ describes two components of the electronic doublet, and $f(\mathbf{n}) = (n_x^2 - n_y^2) f_0(n)$ where $f_0(n)$ is a symmetric function describing the range of the interaction. For simplicity we omit the spin index in the sum. The model could be easily reduced to a lattice gas model. Let us introduce the classical variable $\Phi_{\mathbf{i}} = \langle b_{\mathbf{i}}^+ + b_{\mathbf{i}} \rangle / \sqrt{2}$ and minimize the energy as a function of $\Phi_{\mathbf{i}}$ in the presence of the harmonic term $\omega \sum_{\mathbf{i}} \Phi_{\mathbf{i}}^2 / 2$. We obtain the deformation, which corresponds to the minimum of energy,

$$\Phi_{\mathbf{i}}^{(0)} = -\sqrt{2}g/\omega \sum_{\mathbf{n}} \sigma_{3, \mathbf{i}+\mathbf{n}} f(\mathbf{n}). \quad (2)$$

Substituting $\Phi_{\mathbf{i}}^{(0)}$ into the Hamiltonian (1) and taking into account that the carriers are charged we arrive at the lattice gas model. We use a pseudospin operator $S = 1$ to describe the occupancies of the two electronic levels n_1 and n_2 . Here $S^z = 1$ corresponds to the state with $n_1 = 1$, $n_2 = 0$, $S_i^z = -1$ to $n_1 = 0$, $n_2 = 1$ and $S_i^z = 0$ to $n_1 = n_2 = 0$. Simultaneous occupancy of both levels is excluded due to a high onsite Coulomb repulsion energy. The Hamiltonian in terms of the pseudospin operator is given by

$$H_{LG} = \sum_{\mathbf{i}, \mathbf{j}} (-V_l(\mathbf{i} - \mathbf{j}) S_{\mathbf{i}}^z S_{\mathbf{j}}^z + V_c(\mathbf{i} - \mathbf{j}) Q_{\mathbf{i}} Q_{\mathbf{j}}), \quad (3)$$

where $Q_{\mathbf{i}} = (S_{\mathbf{i}}^z)^2$. $V_c(\mathbf{n}) = e^2/\epsilon_0 a (n_x^2 + n_y^2)^{1/2}$ is the Coulomb potential, e is the charge of electron, ϵ_0 is the static dielectric constant and a is the effective unit cell period. The anisotropic short range attraction potential is given by $V_l(\mathbf{n}) = g^2/\omega \sum_{\mathbf{m}} f(\mathbf{m}) f(\mathbf{n}+\mathbf{m})$. The attraction in this model is generated by the interaction of electrons with optical phonons. The radius of the attraction force is determined by the radius of the electron-phonon interaction and the dispersion of the optical phonons[8].

A similar model can be formulated in the limit of continuous media. In this case the deformation is characterized by the components of the strain tensor. For the two dimensional

case we can define 3 components of the strain tensor: $e_1 = u_{xx} + u_{yy}$, $\epsilon = u_{xx} - u_{yy}$ and $e_2 = u_{xy}$ transforming as the A_{1g} , B_{1g} and B_{2g} representations of the D_{4h} group respectively. These components of the tensor are coupled linearly with the two-fold degenerate electronic state which transforms as the E_g or E_u representation of the point group. Similarly to the case of previously considered interaction with optical phonons we keep the interaction with the deformation ϵ of the B_{1g} symmetry only. The Hamiltonian without the Coulomb repulsion term has the form:

$$H = g \sum_{\mathbf{i}} S_{\mathbf{i}}^z \epsilon_{\mathbf{i}} + \frac{1}{2} (A_1 e_{1,\mathbf{i}}^2 + A_2 \epsilon_{\mathbf{i}}^2 + A_3 e_{3,\mathbf{i}}^2), \quad (4)$$

where A_j are corresponding components of the elastic modulus tensor. The components of the strain tensor are not independent [12, 15] and satisfy to the compatibility condition. The compatibility condition leads to a long range anisotropic interaction between polarons. The Hamiltonian in the reciprocal space has the form:

$$H = g \sum_{\mathbf{k}} S_{\mathbf{k}}^z \epsilon_{\mathbf{k}} + (A_2 + A_1 U(\mathbf{k})) \frac{\epsilon_{\mathbf{k}}^2}{2}. \quad (5)$$

The Fourier transform of the potential is given by:

$$U(\mathbf{k}) = \frac{(k_x^2 - k_y^2)^2}{k^4 + 8(A_1/A_3)k_x^2 k_y^2}. \quad (6)$$

By minimizing the energy with respect to ϵ_k and taking into account the long-range Coulomb repulsion we again derive Eq.(3). The anisotropic interaction potential $V_l(\mathbf{n}) = -\sum_{\mathbf{k}} \exp(i\mathbf{k} \cdot \mathbf{n}) \frac{g^2}{2(A_2 + A_1 U(\mathbf{k}))}$ is determined by the interaction with the classical deformation and is long-range. It decays as $1/r^2$ at large distances in 2D. Since the attraction forces decay faster then the Coulomb repulsion forces at large distances the attraction can overcome the Coulomb repulsion at short distances, leading to a mesoscopic phase separation.

Irrespective of whether the resulting interaction between polarons is generated by acoustic or optical phonons the main physical picture remains the same. In both cases there is an anisotropic attraction between polarons on short distances. The interaction could be either ferromagnetic or antiferromagnetic in terms of the pseudospin operators depending on mutual orientation of the orbitals. Without losing generality we assume that $V(\mathbf{n})$ is nonzero only for the nearest neighbors.

Our aim is to study the model (Eq.(3)) at constant average density,

$$n = \frac{1}{N} \sum_{\mathbf{i}} Q_{\mathbf{i}}, \quad (7)$$

where N is the total number of sites. However, to clarify the physical picture it is more appropriate to perform calculations with a fixed chemical potential first by adding the term $-\mu \sum_i Q_i$ to the Hamiltonian (3).

Models such as (3), but in the absence of the long-range forces, were studied many years ago on the basis of the molecular-field approximation in the Bragg-Williams formalism [16, 17]. The mean-field equations for the particle density n and the pseudospin magnetization $M = \frac{1}{N} \sum_i S_i^z$ have the form[16]:

$$M = \frac{2 \sinh(2zV_l M/k_B T)}{\exp(-\mu/k_B T) + 2 \cosh(2zV_l M/k_B T)} \quad (8)$$

$$n = \frac{2 \cosh(2zV_l M/k_B T)}{\exp(-\mu/k_B T) + 2 \cosh(2zV_l M/k_B T)} \quad (9)$$

here $z = 4$ is the number of the nearest neighbours for a square lattice in 2D and k_B is the Boltzman constant. A phase transition to an ordered state with a finite M may be of either the first or the second order, depending on the value of μ . For $\mu > 0$ it is always of the second order and $n \rightarrow 1$ as $T \rightarrow 0$. For the large negative values $\mu < -2zV_l$ the phase transition is absent and $n \rightarrow 0$ as $T \rightarrow 0$. For the physically important case $-2zV_l < \mu < 0$ ordering occurs as a result of the first order phase transition. Two solutions of Eqs.(8,9) with $M = 0$ and with finite M corresponds to two different minima of the free energy. The line of the phase transition is determined by the condition: $F(M = 0, \mu, T) = F(M, \mu, T)$ where M is the solution of Eq. (8) (Fig.1)). When the number of particles is fixed (Eq.9) the system is unstable with respect to global phase separation below T_{crit} . As a result at fixed average n two phases with $n_0 = n(M = 0, \mu, T)$ and $n_M = n(M, \mu, T)$ coexist.

To investigate further the properties of the system, we performed Monte-Carlo (MC) simulations of the Hamiltonian Eq.(3). The simulations were performed on a square lattice with dimensions up to $L \times L$ sites with $10 \leq L \leq 100$ using a standard Metropolis algorithm[18] in combination with simulated annealing[19].

First, for comparison with MF theory, the Monte-Carlo simulation of the model Eq.(3) in absence of the Coulomb forces shows the reduction of T_{crit} due to fluctuations in 2D, by a factor of ~ 2 (Fig.1).

Next we include the Coulomb interaction $V_c(r)$. We use open boundary conditions to avoid complications due to the long range Coulomb forces and ensure global electroneutrality by adding an electrostatic potential due to the uniformly charged background $V_{jell}(\mathbf{i})$ to (3), such that the Monte Carlo interaction becomes $H_{MC} = H_{LG} + \sum_i Q_i V_{jell}(\mathbf{i})$, with the

total background charge being equal in magnitude to the total charge of carriers, $e \sum_i Q_i$, but with opposite sign. The dimensionless temperature $t = k_B T \epsilon_0 a / e^2$, and the energy per particle are defined as $e_{MC} = H_{MC} \epsilon_0 a / N e^2$. The short range potential $v_l(\mathbf{i}) = V_l(\mathbf{i}) \epsilon_0 a / e^2$ was taken to be nonzero only for $|\mathbf{i}| < 2$ and was therefore specified for nearest neighbours and next nearest neighbours as $v_l(1, 0)$ and $v_l(1, 1)$ respectively.

The results of the Monte Carlo simulations could be summarized as follows. (i) Coulomb repulsion leads to further reduction of the onset temperature of phase separation (see Fig.1). (ii) Depending on the value of the attractive potential and its anisotropy phase separation takes place in the form of bubbles, horizontal and vertical or diagonal stripes (Fig.2). (iii) Depending on the anisotropy of the potential stripes (bubbles) are ferromagnetically or antiferromagnetically orbitally ordered (Fig.2). (iv) The size of clusters is determined by the ratio of short range attraction and long-range Coulomb energy.

The results of the Monte-Carlo simulation of the model (3) allow general model independent interpretation. Let us consider the classical free energy density corresponding to the first order phase transition:

$$F_1 = ((t - 1) + (\Lambda^2 - 1)^2) \Lambda^2 \quad (10)$$

Here t is dimensionless temperature. Let us assume that the order parameter Λ is coupled to the local charge density ρ , $F_{coupl} = \alpha(\Lambda^2 - \rho)^2$. The total free energy density should contain the gradient term $F_{grad} = C(\nabla \Lambda)^2$ and electrostatic energy $F_{el} = K \phi \rho$ as well. The electrostatic potential is determined from the Poisson equation $\epsilon_0 \Delta \phi = 4\pi e(\rho - \rho_0)$. Here we write ρ_0 explicitly to take into account global electroneutrality. Substituting the solution of the Poisson equation to the electrostatic energy we obtain $F_{el} = K' \sum_{\mathbf{k}} V(k)(\rho - \rho_0)_{\mathbf{k}}^2$, here $V(k) = 1/k$. Minimization of $F_1 + F_{coupl} + F_{grad} + F_{el}$ at fixed t and ρ_0 gives the spatial variation of the order parameter. The results of minimization are presented in Fig.3. As clearly seen from Fig.3 at low density phase separation takes place in the form of bubbles of optimal radius which are ordered. When density increases charge segregation appears in the form of charged domain walls.

Let us assume that a single bubble of the ordered phase with the radius R has appeared. As it was discussed in [13, 20] the energy of the bubble is determined as: $\epsilon = -F\pi R^2 + \alpha\pi R + \gamma\pi R^3$, here F is the energy difference between two minima in the free energy, α is the surface energy, and γ determines the energy due local breaking of the charge neutrality.

If $\alpha < \pi F/3\gamma \epsilon$ has well defined minimum. This minimum corresponds to optimal size of the cluster. Residual interactions leads to interbubble interactions and ordering of clusters at low temperatures.

We have demonstrated that anisotropic interaction between Jahn-Teller centers generated by optical and/or acoustical phonons leads to the short scale phase separation in the presence of the long range Coulomb repulsion. Topology of texturing differs from charged bobbles to oriented charged stripes depending on the anisotropy of short range potential.

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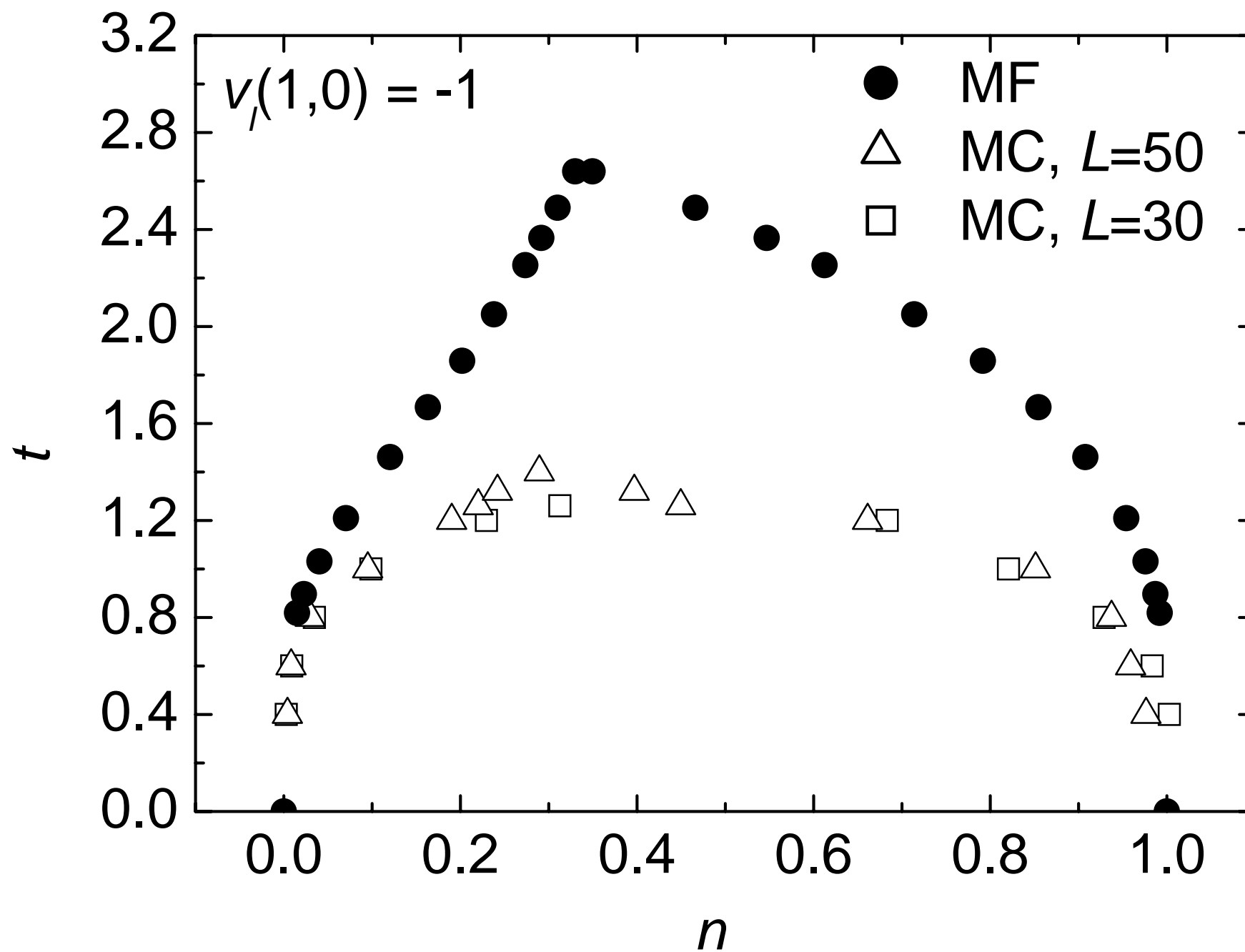
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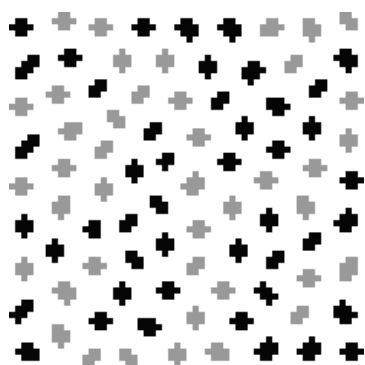
I. FIGURE CAPTIONS

Fig. 1 The phase diagram of the model in absence of the Coulomb repulsion. The full symbols represent the mean field (MF) solution while empty symbols represent the Monte-Carlo (MC) simulation result on systems with two different sizes L .

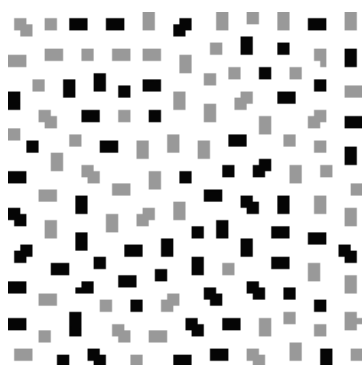
Fig. 2 Snapshots of clusters ordering at $t = 0.04$, $n = 0.2$ and $v_l(1, 0) = -1$ as a function of $v_l(1, 1)$.

Fig. 3 Snapshots of the order parameter distribution at different average densities obtained by minimization of the free energy (10). With increasing of density crossover from charged bubbles to the charged domain walls is clearly observed.

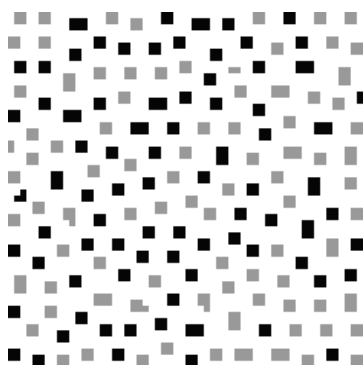




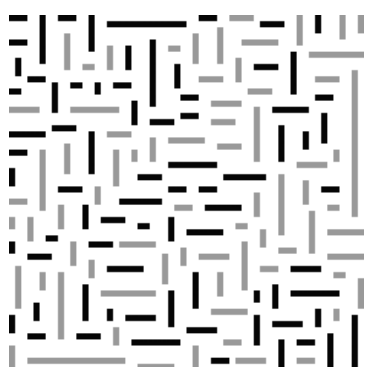
$$v_l(1,1) = -1$$



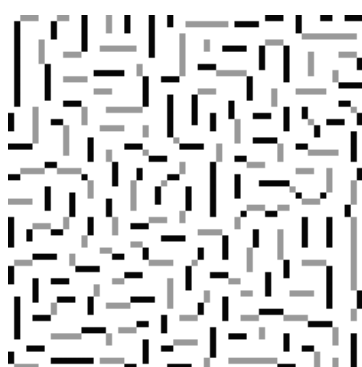
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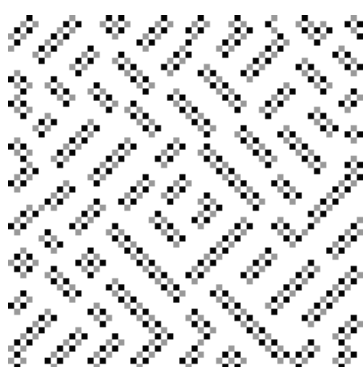
$$v_l(1,1) = -0.2$$



$$v_l(1,1) = 0.2$$



$$v_l(1,1) = 0.5$$



$$v_l(1,1) = 1$$

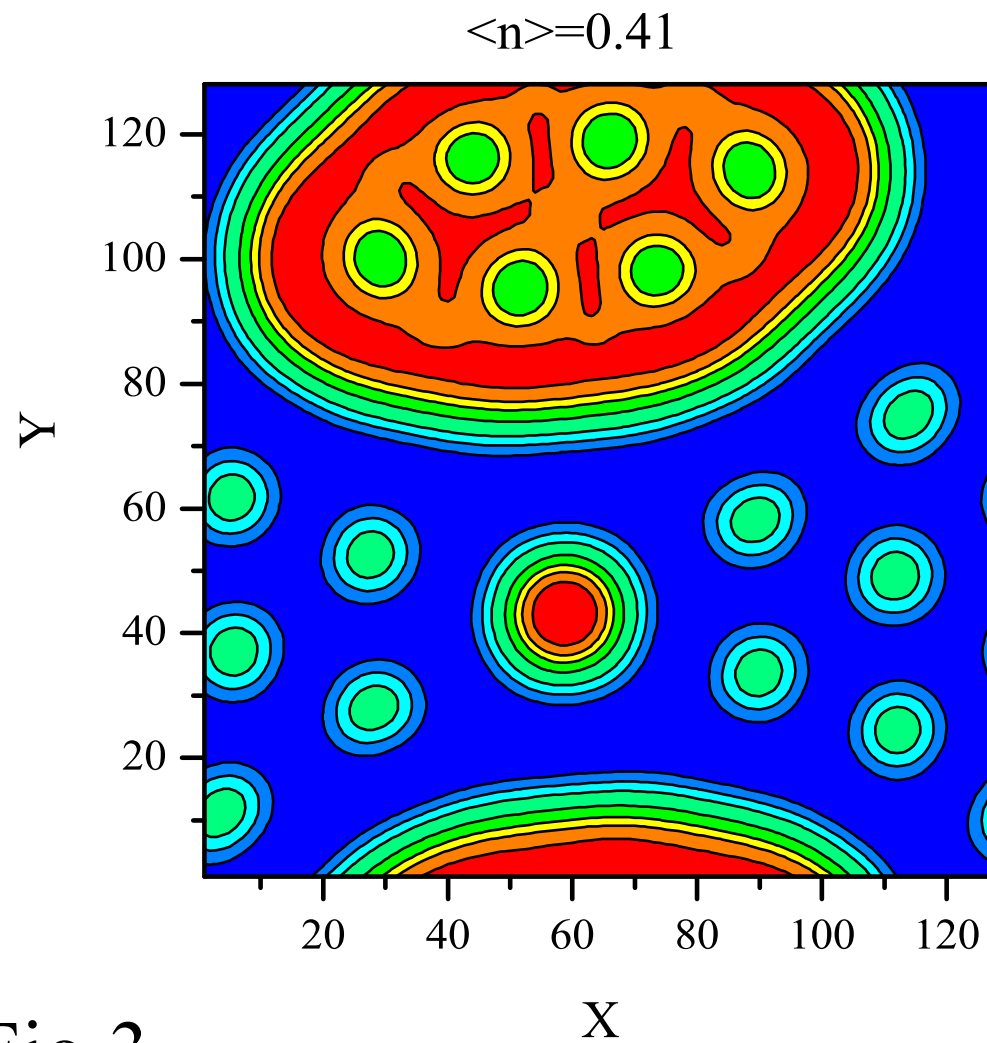
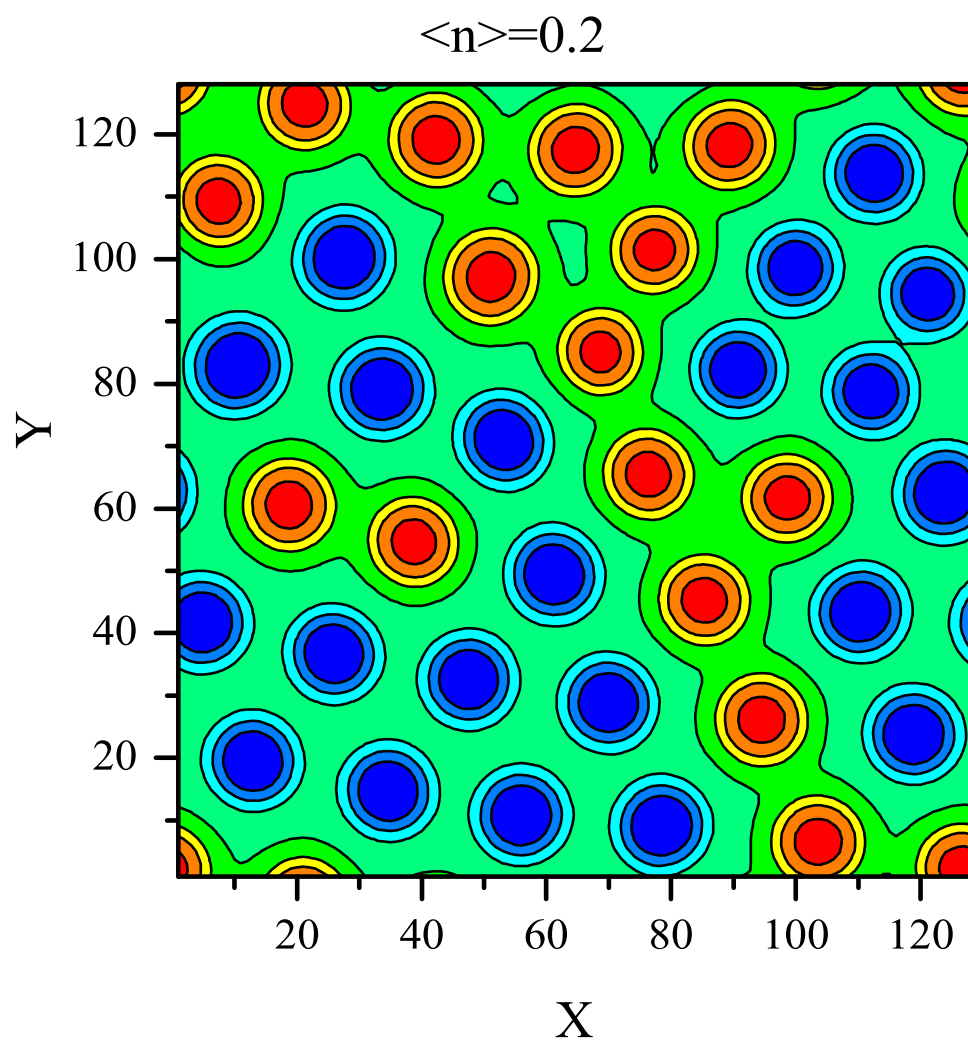


Fig.3